Hidden correlations of exciton complexes in self-assembled quantum dots and rings

Weiwei Zhang, Ming Gong, Chuan-Feng Li, Guang-Can Guo, and Lixin He*

Key Laboratory of Quantum Information, University of Science and
Technology of China, Hefei, 230026, People's Republic of China
(Dated: December 8, 2008)

The binding energies of trions (X^+, X^-) and biexciton (XX) in self-assembled semiconductor quantum dots (QDs) are very sensitive to the geometry and chemical composition of the QDs, and are random from dots to dots. However, in this letter, we show through analytical and numerical methods that the transition energies of the exciton complexes in self-assembled quantum dots and rings follow a simple and robust rule, i.e., the sum of exciton and biexciton transition energies minus the transition energies of trions is always positive and almost a constant for the same type of quantum dots and rings as a consequence of a pure Coulomb correlation effect. More interestingly, this quantity show a sharp transition when the topology change from a dot to a ring. This *hidden* correlation effect, directly measurable in experiments, offers a useful way to understand the photoluminescence spectra of self-assembled quantum dots and rings.

New physics emerge when the size of a solid system reduces to nano-scale. In the self-assembled semiconductor quantum dots (QDs), the three-dimensional confinement effects lead to atom-like electronic structure and long-living coherent quantum states. The confinement also enhances the Coulomb interactions among the quasiparticles, leading to novel physics in QDs other than bulk materials, e.g., Coulomb blockade effects[1], and the non-Aufubau filling order for holes [2, 3] in the InAs/GaAs quantum dots. The unique properties of QDs are not only of special interests in the view of fundamental physics, but also have important applications in quantum information processes. In these applications, the exciton complexes, including exciton (X), biexciton (XX) and trions (X^+, X^-) play extremely important roles. For example, neutral excitons can be used to generate single-photons [4, 5], whereas biexcitons can be used to generate entangled photon pairs [6, 7]. Trions can be used to write in/read out the information of spin qubit, or to manipulate the spin states [8, 9, 10, 11]. Due to the enhanced Coulomb interactions, the transition energies of the biexcitons and trions have significant (a few meV) energy shifts (i.e., binding energy) relative to those of the neutral excitons. Question arise that if we can find some simple relations between the transition energies of these exciton complexes?

Unfortunately, both experimental measurements [12, 13] and theoretical calculations [14] show that the binding energies of the exciton complexes change dramatically, even their signs, with respect to the sizes, shapes and chemical compositions of the QDs. Even worse, the binding energies seems to be random from dot to dot. It seems hopeless to find some simple relations between the transition energies of the exciton complexes. In this letter, we show however, through analytical analysis and numerical calculations that there is indeed a simple and robust relations among the transition energies of the exciton complexes in self-assembled quantum dots and rings.

The binding energies of exciton complexes vary mainly due to the competition of the direct electron-electron, hole-hole and electron-hole Coulomb interactions[12, 14], which strongly depend on the size or the composition of a QD. To eliminates the influence of direct Coulomb interactions, we define a quantity Δ,

$$\Delta = X + XX - X^+ - X^- , \qquad (1)$$

where, X, XX, X^+, and X^- are the transition energies of excitons, biexcitons, positive and negative trions, respectively. It is easy to show that $\Delta=0$ under the Hartree-Fock (or single configuration) approximation, as the direct Coulomb energies and exchange energies cancel each other for the four types of exciton complexes. However, Δ is generally not zero when we include correlation energies.

We have calculated the photoluminescence (PL) spectra for a large amount of QDs with different sizes (radii and heights), compositions (InGaAs/GaAs or InPAs/InP) and shapes (lens, cone, elongated and pyramidal dots). We first obtain the single particle energy levels and wave functions of the geometry-optimized QDs using an empirical pseudopotential method [15], where the total pseudopotential of the system are superposition of the local, screened atomic pseudopotential of all (dot+matrix) atoms and the nonlocal spin-orbit potentials. The pseudopotential Schrödinger equation is solved via the linear combination of bulk bands (LCBB) method [16]. Many-body effects are included via the configuration interaction (CI) method [17] by expanding the total wavefunction in Slater determinants for single and bi-excitons formed from all of the confined single-particle electron and hole states.
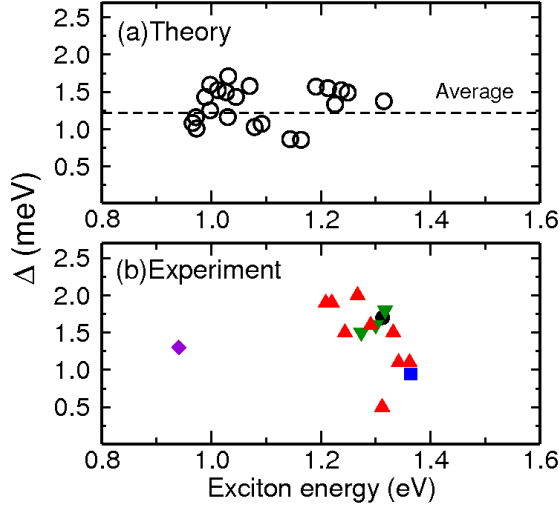
*corresponding author: helx@ustc.edu.cn

FIG. 1: (color online) (a) Calculated Δ for InGaAs/GaAs QDs with various sizes and geometries. (b) Experimental Δ extracted from literatures. (▲), (▼) (●), (♦), and (■) are extracted from Ref. 12, 13, 18, 19, and 20, respectively.
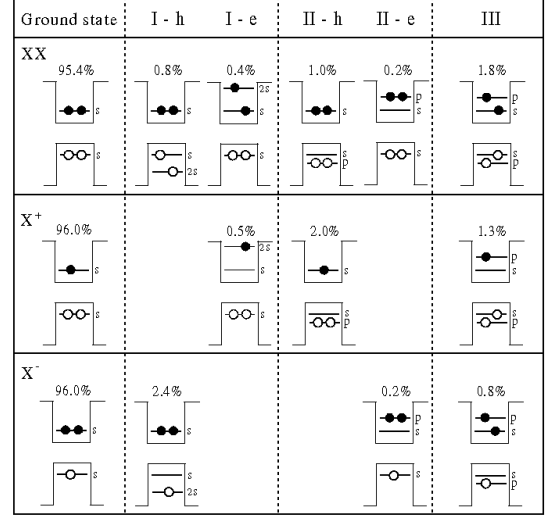

FIG. 2: A schematic figure of the excited configurations that couple substantially to the ground state configurations for XX, X^+ and X^-, divided in three categories (see text for detail). The numbers of percentage in each configuration is the weight of this configuration in the total many-particle wavefunctions from CI calculations. We omit irrelevant single particle levels.

Figure 1(a) depicts the calculated Δ in InGaAs/GaAs QDs with respect to their exciton energies. It clearly shows that Δ, in contrast to the binding energies, are always positive and distributed in a narrow energy range from 0.8 to 1.8 meV even when the exciton energy change dramatically from 0.9 to 1.4 eV, with an average value equal to about 1.22 meV. We see that Δ hardly depends on the size, composition, and even the shape of the QDs. Therefore Δ represent a simple and robust relationship between the transition energies of exciton complexes. To verify the calculations, we show in Fig. 1(b) experimental values of Δ extracted from the InGaAs/GaAs QDs PL spectra in the literatures[12, 13, 18, 19, 20] of four different groups. Indeed, the experimental Δ distributes in 1.0 - 2.0 meV as the exciton energy varies from 0.9 to 1.4 eV, which strongly support the theoretical predictions. We have also calculated the Δ for InAs/InP QDs and find that Δ are distributed in 0.6 - 1.3 meV.

Vanishes in the HF approximation, Δ is therefore a pure correlation effect. It not only represents a simple relation between the transition energies of the exciton complexes, but also provides an easy way to characterize the correlation effects in different nano-structures, which was previously only avaliable in theoretical calculations. In order to gain an analytic understanding of these results, we take use of a perturbation theory. We use a typical lens-shaped InAs/GaAs QD as an example. Other kind of dots can be studied in a similar way. Since $\Delta=0$ under Hartree-Fock approximation, Δ can be alternatively calculated as,

$$\Delta = \Delta_{XX} - \Delta_{X^+} - \Delta_{X^-}, \qquad (2)$$

where, Δ_{X^+}, Δ_{X^-}, and Δ_{XX} are the correlation energies (i.e., the energy difference between CI and HF calculations) for the trions and biexcitions, respectively. For example, $\Delta_{X^+} = E_{CI}(X^+) - E_{HF}(X^+)$. To get the correlation energies for each type of excitons, we first solve the many-particle Hamiltonian in a single configuration approximation and obtain the energy $E_j^{(HF)}$ for j-th configuration $|\Phi_j\rangle$. To the second order approximation, the correlation energy of an exciton is,

$$\Delta_X = \sum_{j\neq 0} \frac{|\langle \Phi_0(X)|H_I|\Phi_j(X)\rangle|^2}{E_0^{(HF)}(X) - E_j^{(HF)}(X)} \qquad (3)$$

where, H_I is the Coulomb interactions among the quasiparticles. $|\Phi_0(X)\rangle$ and $E_0^{(HF)}(X)$ are the ground state configuration and its energy for an exciton. The correlation energies of XX, and trions X^+, X^- can be calculated in the exact same procedures.

We show schematically in Fig. 2 the excited configurations that couple substantially to the ground states of XX, X^+ and X^-. These configurations can be classified into three categories. We define the configurations in which only one electron (or hole) is excited as type I configurations. Type II configurations are those that two electrons (or two holes) are simultaneously excited, whereas type III configurations contain one excited electron and one excited hole. Configurations of more than two particle excitations do not couple to the ground state.

Type I configurations can be further divided as electron excitations (type I-e) and hole excitations (type I-

h). As we see from Fig. 2, for X^+ only type I-e excitations contribute significantly to the correlation energy, whereas for X^-, only type I-h excitations contribute significantly. On the other hand, both I-e and I-h contribute to the correlation energies of biexcitons. The off-diagonal (OD) matrix elements of I-h excitation for XX is $(H_{I-h})_{XX} = 2\langle e_s, h_s; e_s, h_{2s}\rangle + \langle h_s, h_s; h_s, h_{2s}\rangle$ and for X^- is $(H_{I-h})_{X^-} = 2\langle e_s, h_s; e_s, h_{2s}\rangle$. Here, we used a short notation, $\langle e_s, h_s; e_s, h_{2s}\rangle =$

$$\int\int \frac{\phi_{e_s}^*(\boldsymbol{r}_1)\phi_{h_s}^*(\boldsymbol{r}_2)\phi_{e_s}(\boldsymbol{r}_1)\phi_{h_{2s}}(\boldsymbol{r}_2)}{\epsilon(\boldsymbol{r}_1 - \boldsymbol{r}_2)|\boldsymbol{r}_1 - \boldsymbol{r}_2|} d^3\boldsymbol{r}_1 d^3\boldsymbol{r}_2\,. \quad (4)$$

where, $\phi_{e_s}(\boldsymbol{r}_1)$, $\phi_{h_s}(\boldsymbol{r}_2)$, $\phi_{h_{2s}}(\boldsymbol{r}_2)$ are the electron (e) and hole (h) single particle wavefunctions, and $\epsilon(\mathbf{r}_1 - \mathbf{r}_2)$ is the dielectric function [17]. For simplicity, we dropped the spin notation in the above equation, but included it in real calculations. Since electron and hole wavefunctions have similar dimensions in QDs [15], $\langle e_s, h_s; e_s, h_{2s}\rangle \sim$ - $\langle h_s, h_s; h_s, h_{2s}\rangle$. Therefore $\langle h_s, h_s; h_s, h_{2s}\rangle$ almost cancels $\langle e_s, h_s; e_s, h_{2s}\rangle$. As a result, $(H_{I-h})_{XX}$ is much smaller than $(H_{I-h})_{X^-}$. This cancellation effect is also the reason that I-e excitations contribute little to the correlation energy of X^+. For the other I-h configurations, the OD term vanishes in cylindrical dots , because $\langle e_s, h_s; e_s, h_p\rangle$, $\langle e_s, h_s; e_s, h_d\rangle = 0$ due to the symmetry of the wavefunctions.

The difference of the denominators of in calculating the correlation energies of XX and X^- [see Eq. (3)] also contributes to Δ. The single-particle part of the denominators are the same for XX and X^-, and the difference comes from many-particle interactions. It turns out that for XX and X^-, the difference between the two denominators is $J_{ss}^{hh} - J_{s-2s}^{hh}$, i.e., the difference of Coulomb energies between the hole s state with itself and s with $2s$ states. This energy difference is much smaller than the denominators themselves. The correlation energy of XX from I-h excitations has a additional factor of 2 due to spin degeneracy. Combining all above factors, we find that I-h contribution to the correlation energies $|\Delta_{X^-}^{I-h}|$ is much larger than $|\Delta_{XX}^{I-h}|$. Using the numerically calculated Coulomb integrals and single particle levels, for a lens-shaped dot with base D=25 nm, height h=2 nm, $\Delta_{X^-}^{I-h} \approx 2.5\ \Delta_{XX}^{I-h}$. The I-h configurations' contribution to Δ is about 0.93 meV.

We can apply similar analysis to type I-e configurations, and find they also make positive contribution to Δ. However, the contribution from I-e excitations are much smaller than those of I-h ones, because electrons have much larger level spacing than that of holes in the InAs/GaAs dots and therefore larger denominators (about 3 times) in Eq. 3 than those I-h excitations. For the same dot above, I-e configurations contribute 0.31 meV to Δ.

Type II configurations can also be sub-classified into II-h and II-e excitations. The II-h OD matrix elements for XX are $(H_{II-h})_{XX} = \langle h_s, h_s; h_i, h_j\rangle$, where i, j are the indices of the excited single-particle levels. If $i \neq j$, the configurations would not couple to the ground state configuration in cylidrical dots. For $i=j$ configurations, the most contributions are from p orbitals excitions. The contributions from higher energy excitions are much smaller due to larger energy spacing. Interestingly, we find that X^+ has the same OD matrix elements for II-h excitations i.e. $(H_{II-h}^{OD})_{X^+} = (H_{II-h}^{OD})_{XX}$. As a result, only the difference between the denominators contributes to Δ. For type II-h configurations, the difference between two denominators $\sim 2(J_{ss}^{eh} - J_{si}^{eh})$. We find that $\Delta E_{II-h}^{\mathrm{HF}}(XX)$ is always larger than $\Delta E_{II-h}^{\mathrm{HF}}(X^+)$, because the coulomb integrals always satisfy $J_{ss}^{eh} > J_{si}^{eh}$. As a result, type II-h configurations always make positive contribution to Δ, but usually much smaller than those of type I configurations. Similar analysis applies for type II-e configurations. The denominator $\Delta E_{II-e}^{\mathrm{HF}}(XX) > \Delta E_{II-e}^{\mathrm{HF}}(X^-)$. Therefore, type II-e configurations also make positive contribution to Δ. However, due to much larger single particle level spacing of electrons, the contribution from type II-e configurations is even smaller than that from type II-h ones.

Type III configurations contain one excited electron and one excited hole, as shown in Fig. 2. We find that the OD Hamiltonian matrix elements for for XX, X^+ and X^- are the same for each type of excitation, i.e., $(H_{III}^{OD})_{XX} = (H_{III}^{OD})_{X^+} = (H_{III}^{OD})_{X^-} = \langle e_s, h_s; e_i, h_j\rangle$, where i, j are the indeces of the excited single particle levels in the configurations. The denominators in calculating the correlation energies for these three types of exciton complexes [see Eq. (3)] are also very close for type III configurations. After consider the spin degeneracy, the correlation energy of XX almost cancels those of trions, and type III configurations have very little contribution to Δ.

For typical cylindrical dots, the hidden correlation Δ mostly comes from type I configurations. The type II contribution is less but significant, whereas the type III contribution is negligible. For example, for the lens-shaped dot with D=25 nm and h=2 nm, the perturbation theory gives Δ=1.51 meV, in which type I configurations contribute 1.23 meV and type II ones contribute the remainder 0.28 meV. As to the individual configurations, the type I hole $2s$ excitation have the most significant contributions to Δ. This non-trivial result suggests that not including the $2s$ orbital in the CI basis will cause huge error in calculating Δ. The perturbation theory gives a little smaller Δ than that from CI method (1.80 meV), due to ignoring higher order terms. We have applied the same analysis for other type dots, with different sizes and shapes (e.g. elongated dots and pyramidal dots). Even though the details of the contributions from each configuration may be quite different, the final results of Δ are very close, which suggests that there are more fundamen-
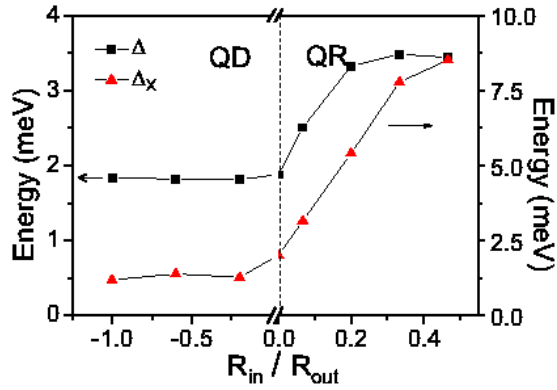
FIG. 3: The correlation energy of the exciton Δ_X and the *hidden* correlation Δ as functions of R_{in}/R_{out} in a InAs/GaAs nanostructure. When R_{in}/R_{out} <0, the nanostructure is a QD, and becomes a QR when R_{in}/R_{out} >0.

tal physics remain to be resolved in future studies.

We now discuss another type of important semiconductor nano-structures, quantum rings (QRs), which have non-simply connected topology. In a QR, the single-particle energy level is more sensitive to the inner radii of the ring, whereas the Coulomb interactions dependent more strongly on the outer radii of the rings [21, 22]. The single-particle energy levels in a QR are much more condensed than in a QD [21, 22] of similar sizes, and therefore the particles are expected to be more correlated. Figure 3 depicts the correlation energy of an exciton Δ_X and Δ with respect to the ratio of inner radius to outer radius (R_{in}/R_{out}) in a InAs/GaAs QR. We adapt the ring model from Ref. 22. The outer radius of the ring is fixed at R_{out}= 15 nm, whereas the inner radius R_{in} increase from -15 nm to 7 nm. The structure with $R_{in} < 0$ is a QD and the structure with $R_{in} > 0$ is a QR. Figure 3 shows that when the structure changes from a QD to a QR, Δ_X increases dramatically from about 2 meV to about 10 meV. Remarkably, almost a constant in the QD region, Δ shows a sharp transition when the topology changes from a dot to a ring, where Δ jumps from 1.8 meV in the QD region to 3.5 meV in the QR region and saturates when R_{in}/R_{out} exceeds 0.2. This effect can be directly measured in a dot to ring transition experiments [23].

To conclude, we have introduced a *hidden* correlation function as the sum of exciton and biexciton transition energies minus the transition energies of trions for the exciton complexes in semiconductor nanostructures such as quantum dots and rings. Measurable in experiments, the hidden correlation provides a deep insight to the Coulomb correlation effects of the exciton complexes. We show that the hidden correlation energy is positive and almost a constant for the same type of quantum dots. Remarkably, it shows a sharp phase-transition-like behave

when the topology changes from a dot to a ring. The fundamental physics that govern this intriguing behaver remain to be fully explored.

[1] M. Field, C. G. Smith, M. Pepper, D. A. Ritchie, J. E. F. Frost, G. A. C. Jones, and D. G. Hasko, Phys. Rev. Lett **70**, 1311 (1993).
[2] L. He, G. Bester, and A. Zunger, Phys. Rev. Lett. **95**, 246804 (2005).
[3] D. Reuter, *et al.*, Phys. Rev. Lett. **94**, 026808 (2005).
[4] Z. Yuan, B. E. Kardynal, R. M. Stevenson, A. J. Shields, C. J. Lobo, K. Cooper, N. S. Beattie, D. A. Ritchie, and M. Pepper, Science **295**, 102 (2002).
[5] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, and A. Imamoğlu, Science **290**, 2282 (2000).
[6] N. Akopian, N. H. Lindner, E. Poem, Y. Berlatzky, J. Avron, D. Gershoni, B. D. Gerardot, and P. M. Petroff, Phys. Rev. Lett. **96**, 130501 (2006).
[7] R. M. Stevenson, R. J. Young, P. Atkinson, K. Cooper, D. A. Ritchie, and A. J. Shields, Nature **439**, 179 (2006).
[8] J. Berezovsky, M. H. Mikkelsen, O. Gywat, N. G. Stoltz, L. A. Coldren, and D. D. Awschalom, Science **314**, 1916 (2006).
[9] J. Berezovsky, M. H. Mikkelsen, N. G. Stoltz, L. A. Coldren, and D. D. Awschalom, Science **320**, 349 (2008).
[10] D. Press, T. D. Ladd, B. Zhang, and Y. Yamamoto, Nature **456**, 218 (2008).
[11] B. D. Gerardot, D. Brunner, P. A. Dalgarno, P. Öhberg, S. Seidl, M. Kroner, K. Karrai, N. G. Stoltz, P. M. Petroff, and R. J. Warburton, Nature **451**, 441 (2008).
[12] S. Rodt, A. Schliwa, K. Pötschke, F. Guffarth, and D. Bimberg, Phys. Rev. B **71**, 155325 (2005).
[13] R. Seguin, A. Schliwa, T. D. Germann, S. Rodt, K. Pötschke, A. Strittmatter, U. W. Pohl, and D. Bimberg, Appl. Phys. Lett **89**, 263109 (2006).
[14] G. Bester and A. Zunger, Phys. Rev. B **68**, 073309 (2003).
[15] A. J. Williamson, L.-W. Wang, and A. Zunger, Phys. Rev. B **62**, 12963 (2000).
[16] L.-W. Wang and A. Zunger, Phys. Rev. B **59**, 15806 (1999).
[17] A. Franceschetti, H. Fu, L.-W. Wang, and A. Zunger, Phys. Rev. B **60**, 1819 (1999).
[18] P. A. Dalgarno, J. McFarlane, B. D. Gerardot, and R. J. Warburton, Appl. Phys. Lett **89**, 043107 (2006).
[19] N. I. Cade, H. Gotoh, H. Kamada, T. Tawara, T. Sogawa, and H. Nakano, Appl. Phys. Lett **87**, 172101 (2005).
[20] A. J. Shields, Nature Photonics **1**, 215 (2007).
[21] S.-S. Li and J.-B. Xia, J. Appl. Phys. **89**, 3434 (2001).
[22] W. Zhang, Z. Su, M. Gong, C. Li, G. Guo, and L. He, Europhys. Lett **83**, 67004 (2008).
[23] D. Granados and J. M. Garcia, Appl. Phys. Lett. **82**, 2401 (2003).